Splitting of the superconducting transition and the magnetism in UPt_3

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This paper discusses a possible reason for splitting in the superconducting transition in UPt_3, which may result from the fairly strong coupling of conduction electrons with the magnetic moments of uranium atoms. This assertion is based on a simple model described by a Ginzburg–Landau functional with a two-component order parameter $\boldsymbol{\psi}$. The Ginzburg–Landau functional without interaction possesses the complete symmetry of a hexagonal crystal, D_{6h}. Uranium atoms, however, have magnetic moments $\mathbf{M}$, and the interaction term $\gamma|\mathbf{M}\boldsymbol{\psi}|^2$ in the Ginzburg–Landau functional breaks this symmetry locally. Averaging over the various configurations of the vector $\mathbf{M}$ is performed with allowance for the finite correlation radius a, on the assumption $a < \xi$, with ξ the coherence length. This restores the symmetry of the effective Ginzburg–Landau functional up to D_{6h}, which means that in a real crystal the hexagonal symmetry is not broken on a scale larger than ξ. Within this theory expressions for the discontinuities in the specific heat and an equation linking the upper critical field H_{c2} with the splitting ΔT_c of the phase transition under pressure variations are obtained. The difficulties associated with the fact that the magnetic moments of uranium atoms are small are discussed.

1. INTRODUCTION

Recent years have seen a surge in experimental work supporting the idea that there is splitting in the superconducting phase transition in UPt_3 (Refs. 1–3). The temperature dependence of the upper critical field H_{c2} also exhibits a fairly sharp break, which suggests the existence of at least two superconducting phases in UPt_3 (Ref. 3). The number of theoretical papers devoted to explaining the experimental data is on the rise.[4–6]

Theories based on the competition among the superconducting states belonging to different representations of the symmetry group D_{6h} of the UPt_3 crystal[4] are satisfactory in that the upper critical field is isotropic in the basal plane, but they do have an important drawback: they cannot explain the relatively low value of the splitting ΔT_c (there are no apparent reasons why the coefficients in the Ginzburg–Landau functional belonging to different representations must be close to each other).

Another large class consists of theories operating within the framework of a single two-dimensional representation of the D_{6h} group.[5,6] In these theoreis the field H_{c2} is generally anisotropic in the basal plane when there is magnetic ordering, say, antiferromagnetism or spin-density waves. This drawback is not insurmountable, since if we consider a mode in which the magnetic-correlations radius a is much smaller than the coherence length ξ, the free energy undergoes self-averaging on a scale greater than ξ, which means there are no preferable directions and isotropy is restored.

Here the way in which this averaging is carried out is very important. A possible approach is the one introduced by Imry and Ma.[7] Theories of this type[4] do not allow for temperature fluctuations (i.e., we have $T = 0$), and in addition a fairly strong assertion is made that the solution for the superconducting order parameter ψ superposed on the given magnetic-moment distribution has the form of a function possessing a characteristic scale L determined by minimizing the free energy as a function of L.

This statement is, to say the least, debatable. The question of the existence of a state of the superconducting-glass type in the given conditions remains open. But, most important, the size of the energy gain obtained in such a theory,

$$\Delta E \sim \gamma M^2 \left(\frac{\gamma M^2 a^2}{\xi_0{}^2} \right)^3$$

is too small. Here γM^2 characterizes the strength of the interaction of $\mathbf{M}$ with ψ (see above), since both a/ξ and γM^2 are small (for $\gamma M^2 \sim 1$, superconductivity is suppressed).

The above reasoning shows that the observable splitting cannot be explained by such effects in view of their smallness; more than that, these effects can be completely ignored.

The theory suggested here also assumes that UPt_3 undergoes a phase transition to a superconducting state whose order parameter transforms according to one of the two-dimensional representations of the hexagonal symmetry group, D_{6h}. In this case, as shown by Volovik and Gor'kov,[8] the density of the Ginzburg–Landau functional without any interaction with $\mathbf{M}$ has the following form

$$F_0 = -\alpha(1 - T/T_c)|\boldsymbol{\psi}|^2 + \beta_1|\boldsymbol{\psi}|^4 + \beta_2|\boldsymbol{\psi}^2|^2, \qquad (1)$$

where $\boldsymbol{\psi} = (\psi_1, \psi_2)$ is a two-dimensional complex-valued vector. The third term on the right-hand side of (1) determines the specific form of the superconducting state: for $\rho_2 > 0$ the solution $\boldsymbol{\psi}^2 = 0$, that is, $\boldsymbol{\psi} = (1,i)$, is preferable energywise; but for $\beta_2 < 0$ the desired state is $\boldsymbol{\psi} = (1,0)$.

The functional (1) describes a "pure" superconductor; from the standpoint of phenomenological theory, the anisotropic interaction of conduction electrons with magnetic moments (as in UPt_3) or with impurities (as in $U_{1-x}Th_xBe_{13}$) is described by a term of the form $\gamma|\mathbf{M}\boldsymbol{\psi}|^2$ in the Ginzburg–Landau functional. The effective free-energy functional can be found by averaging the Ginzburg–Landau functional over the orientations of $\mathbf{M}$.

The possible phase transitions caused by coupling with $\mathbf{M}$ are discussed below, where an expression for some physical quantities is also derived.

2. AVERAGING OVER ORIENTATIONS OF VECTOR M. THE EFFECTIVE FREE-ENERGY FUNCTIONAL

Let us consider a superconductor described by the functional

$$F = \int dV \left[-\alpha \left(1 - \frac{T}{T_c}\right) |\psi(\mathbf{r})|^2 + \beta_1 |\psi(\mathbf{r})|^4 + \beta_2 |\psi^2(\mathbf{r})|^2 + \gamma |\mathbf{M}(\mathbf{r})\psi(\mathbf{r})|^2 \right]. \quad (2)$$

The vector $\mathbf{M}(\mathbf{r})$, as noted earlier, represents the magnetic moment of uranium atoms. It is known[9] that in UPt_3 below $T_N = 5$ K antiferromagnetic order sets in. Hence, it is natural to assume that the vector $\mathbf{M}$ is distributed in direction with a finite correlation radius a:

$$\langle M_i(\mathbf{r}) M_j(\mathbf{r}') \rangle = {}^1/_2 M^2 \delta_{ij} \exp(|\mathbf{r}-\mathbf{r}'|/a), \quad (3)$$

where

$$\mathbf{M}(\mathbf{r}) = (M_1(\mathbf{r}), M_2(\mathbf{r})), \ |\mathbf{M}(\mathbf{r})| = M.$$

To obtain the effective free-energy functional we must average over the orientations of $\mathbf{M}$. Since this averaging has been carried out in Ref. 10, we will not repeat it here. Calculations show that allowing for a finite (nonzero) correlation length a changes the expression for $F_{\rm imp}$ derived in Ref. 10 in the following way:

$$F_{imp} = \frac{\gamma M^2}{2} |\psi|^2 - \frac{1}{\beta a^3} \ln I_0\left(\beta a^3 \frac{\gamma M^2}{2} |\psi^2|\right) \quad (4)$$

where $\beta = T^{-1}$, and

$$I_0(x) = \frac{1}{\pi} \int_0^\pi \exp(x \cos\varphi)\, d\varphi$$

is the zeroth-order modified Bessel function, whose logarithm exhibits the following asymptotic behavior:

$$\ln I_0(x) \approx \begin{cases} x^2/4, & x \to 0, \\ x, & x \to \infty. \end{cases} \quad (5)$$

Note that the case $a \sim a_{\rm U\text{-}U}$, where $a \sim a_{\rm U\text{-}U}$ is the distance between neighboring uranium atoms, has been considered in Ref. 10.

One easily notices that the first term in Eq. (4) only lowers the temperature of the superconducting transition. The effect of the second term is not as clear. If a is fairly small (so small that the independent variable of I_0 is small), the second term in (4) may result in a variation in β_2 in (1). Generally $\beta_2 > 0$ (this follows, for instance, from the weak binding approximation), and if the coupling is fairly strong, the coefficient of the term proportional to $|\psi|^2$ that allows for the interaction, $\beta_{2\,\rm eff}$, $|\psi^2|^2$ may change sign:

$$\beta_{2,\rm eff} = \beta_2 - \frac{\beta a^3}{4}\left(\frac{\gamma M^2}{2}\right)^2. \quad (6)$$

Then, as in the "pure" case, we have $\psi = (1,i)$ for $\beta_{2\,\rm eff} > 0$, but $\psi = (1,0)$ for $\beta_{2\,\rm eff} < 0$, where in contrast to the "pure" case there is another phase transition, at $T_c^{(2)}$. The reason is that the function

$$\tilde{F}_{imp}(|\psi^2|) = \beta_2 |\psi^2|^2 - \frac{1}{\beta a^3} \ln I_0\left(\frac{\gamma M^2}{2} |\psi^2| \beta a^3\right)$$

has a minimum if $\beta_{2\,\rm eff}$ is negative, which follows from the fact that

$$\tilde{F}_{imp}(x) = 0, \ x = 0,$$
$$\tilde{F}_{imp}(x) < 0, \ x > 0 \ (x \to 0),$$
$$\tilde{F}_{imp}(x) > 0, \ x \to \infty.$$

Let $\tilde{F}_{\rm imp}$ have a minimum at point $x = x_0$. Then, as the temperature is lowered, there first occurs a transition to state $\psi = (1,0)$ and then at the point $|\psi|^2 = x_0$ there is a transition to the phase $\psi = (1, i\alpha)$ $(0 < \alpha < 1)$ such that $|\psi^2| = x_0$ (is temperature independent), with $|\psi|^2$ increasing (cf. Ref. 10). If a is fairly large, we have $\beta_{2\,\rm eff} < 0$ in any case (see Ref. 8), which means that splitting of the phase transition does indeed occur. Note that in contrast to the theory of Machida, Ozaki, and Ohni,[6] splitting disappears for a finite value of M, with the exception of the region of extremely high values of a.

Note that as $a \to \infty$ (i.e., in the single-domain case), $F_{\rm imp}(|\psi|)$ tends to zero. This means, as can easily be seen, that superconductivity is not suppressed.

Here are expressions for the jumps in specific heat:

$$\Delta C^+ = \frac{\alpha^2}{2(\beta_1 + \beta_{2\,\rm eff})}, \qquad \Delta C^- = \frac{\alpha^2}{2(\beta_1 + \beta_{2\,\rm eff})} \frac{\beta_{2\,\rm eff}}{\beta_1}, \quad (7)$$

where the subscripts "+" and "−" refer to the "upper" and "lower" transitions, respectively. Note that as $\beta_{2\,\rm eff}$ varies (for instance when pressure changes; see below), the sum of these jumps, $\Delta C^+ + \Delta C^- = \alpha^2/2\beta_1$, is constant, which can be observed in experiments.

According to a recent report,[11] an increase in pressure reduces the splitting in the transition,

$$\frac{d\Delta T_c}{dp} = \frac{d}{dp}(T_c^+ - T_c^-) = -19 \ \text{mK}\cdot\text{kbar}^{-1},$$

and it vanishes at $p^* = 3.7$ kbar and $T^* = 419$ mK. The kink in the temperature dependence of H_{c2} also disappears:

$$\frac{d}{dp}(H_c^+ - H_c^-) = 0.5 \ \text{kOe}\cdot\text{kbar}^{-1}.$$

In this theory, for large values of a we have

$$\frac{T_c^+ - T_c^-}{T_c^+} = \frac{\gamma M^2}{2\alpha}, \quad (8)$$

that is, the decrease in splitting is caused by the weakening in the coupling, which in turn leads to a decrease in $H_c^+ - H_c^-$, specifically

$$\frac{\frac{d}{dp}(T_c^+ - T_c^-)}{\frac{d}{dp}(H_c^+ - H_c^-)} \frac{dH_c^+}{dT} = 1 + \frac{\beta_1}{\beta_2}. \quad (9)$$

In deriving this formula we assumed that the coefficients of the "pure" Ginzburg–Landau functional change with increasing pressure much more slowly than the interaction term γM^2. From (9) it follows that

$$\frac{\beta_1}{\beta_2} \approx 1{,}5\text{–}2{,}0 > 1 \qquad \left(\frac{dH_c^+}{dT} = 63 \ \text{kOe}\cdot\text{K}^{-1}\right),$$

which generally agrees with measured values of specific heat jumps ($\beta_1/\beta_2 \approx 2$–3).

3. CONCLUSION

We have shown that in superconductors in which there is nontrivial pairing (with a two-component order parameter) and a fairly strong anisotropic interaction (with the magnetic moments of the uranium atoms in UPt_3), the superconducting transition splits: as the temperature drops, there first occurs a transition from the normal state to the superconducting phase 1($\psi = (1,0)$) and then a transition from phase 1 to phase 2($\psi = (1,i\alpha)$). The jumps in specific heat in such transitions are comparable.

The intrinsic moment of a Cooper pair in phase 1 is zero while in phase 2 it is nonzero (the magnetic moment of the pair is $\boldsymbol{\mu} \sim i[\boldsymbol{\psi}\boldsymbol{\psi}^*]$, and in phase 1 we have $\boldsymbol{\psi} = \boldsymbol{\psi}^*$). The upper critical field H_{c2} is isotropic in the basal plane, which also agrees with the experimental data.

Of course, within the framework of this theory it is impossible to obtain numerical results. For this one must carry out a consistent microscopic derivation of the interaction term $\gamma|\mathbf{M}\boldsymbol{\psi}|^2$ and perform the averaging over vector $\mathbf{M}$.

There is also a difficulty related to the fact that the observed magnetic moment is extremely small ($\mu \sim 0.01\mu_B$ per uranium atom). As it happens, $\Delta T_c/T_c \sim \gamma M^2/\alpha$, and if μ is very small, $\Delta T_c/T_c$ cannot be of order 0.1, which is observed in experiments (if μ is of order μ_B, then $\gamma M^2/\alpha \sim 1$). A possible explanation of this contradiction consists in the following. Consider the correlator

$$K_{\alpha\beta}(\mathbf{r}\mathbf{r}'t) = \langle \hat{M}_\alpha(\mathbf{r}, 0)\hat{M}_\beta(\mathbf{r}'t)\rangle.$$

where $\hat{M}_\alpha(\mathbf{r})$ is the operator of the magnetic moment of a uranium atom at site $\mathbf{r}$. We take its frequency representation, $K_{\alpha\beta}(\mathbf{r}\mathbf{r}'\omega)$. Since the magnetic transition occurs at $T_N = 5$ K (and $T_c = 0.5\ \mathrm{K} \ll T_N$), we assume that the K correlator will be the following:

$$K_{\alpha\beta}(\mathbf{r}\mathbf{r}'\omega) = M_\alpha(\mathbf{r})M_\beta(\mathbf{r}')\exp(-|\mathbf{r}-\mathbf{r}'|/a)\delta(\omega);$$

where $\mathbf{M}$ is the observed lattice magnetization vector (with $\mathbf{M}(\mathbf{r}) = -\mathbf{M}(\mathbf{r}')$ if $\mathbf{r}$ and $\mathbf{r}'$ are the position vectors of two neighboring uranium atoms). Clearly, K does not vanish at nonzero frequencies. Hence, we must allow for the contributions from these frequencies in the interaction term. Indeed, if a frequency ω is low compared to T_c, the vector $\mathbf{M}$ is unable to average over times τ of order $1/T_c$, and in experiments in measuring the magnetic moments the measurement time τ_m is much longer than $1/T_c$, which means that only the fraction of correlator K up to frequencies $\omega \sim 1/\tau_m \ll T_c$ gets measured. This viewpoint is supported by the fact that in neutron-scattering experiments the size of the correlator is not at all small (approximately corresponding to the magnetic moment $\mu \sim \mu_B$ per uranium atom), and moreover, the correlations become ferromagnetic.[12]

Of course, the above reasoning cannot serve as a rigorous substantiation of the validity of the given model.

Another (but similar) splitting mechanism is also possible. In the presence of shortwave ($k \sim 2k_F$) low-frequency lattice vibrations and nontrivial pairing ($\boldsymbol{\psi}$ is a vector) we return to the results of Ref. 10 because in this situation these soft phonons can be considered as anisotropic impurities (the frequency ω of these phonons is much lower than T_c). In addition, the large effective electron mass ($m^* \sim 10m_e$) may indicate a large coupling constant for the electron–phonon interaction in the range of these vibrations, which means that the interaction term in (2) is fairly large (but here vector $\mathbf{M}$ corresponds to these vibrations rather than to the magnetic moment). The question of which of these two mechanisms is responsible for the splitting in the superconducting transition in UPt_3 (low-frequency phonons or magnetism) remains open.

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